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Tertiary migmatic pegmatites in the Central Rhodope crystalline complex. Uranium-lead zircon dating

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В. Арнаудов, Б. Амов, Ц. Балджиева, М. Павлова. *Третичные мигматитовые пегматиты в кристаллическом комплексе Центральных Родоп. Уран-свинцовая цирконометрия.* Образование послойных мигматитовых пегматитов в принимаемом за докембрийский метаморфическом комплексе у р. Выча южнее г. Кричим, Центральные Родопы, связано с ультраметаморфическими процессами, протекшими в альпийское время. U-Pb возраст цирконов из богатых цирконом и ортитом мигматитовых пегматитов, полученный в двух различных радиогеохронологических лабораториях — в Софии (Болгария) и Амстердаме (Голландия), позднеальпийский — 58-49 Ма. Эти величины близки к определениям, полученным по U-Pb методу для уранинитов из редкометалльных пегматитов (53-49 Ма), секущих сильнометаморфизованные породы в окрестностях с. Долен Златоградского района. Анализ минералогических, геохимических и радиоизотопных данных о послойных и секущих пегматитах и метаморфитах т. наз. Родопской супергруппы не подкрепляет идеи о наличии докембрийских ультраметаморфических пород в Центральных Родопах.

Abstract. The formation of bedded migmatic pegmatites in the presumably Precambrian high-metamorphic complex along the Vâcha River south of the town of Krichim in the Central Rhodope Mts is the result of ultrametamorphic processes which took place in Alpine times. The U-Pb age of zircons from zircon- and allanite-rich migmatic pegmatites is Young Alpine, 58-49 Ma, according to analyses in two radiogechronological laboratories, one in Sofia, Bulgaria, and the other in Amsterdam, Holland. These values are close to the U-Pb ages obtained for uraninites from rare-metal pegmatites (53-49 Ma) crosscutting the high-metamorphic series near the village of Dolen, Zlatograd district. The mineralogical, geochemical and radioisotope evidence gained on the bedded and cross pegmatites in the metamorphic rocks of the so-called Rhodope Supergroup is inconsistent with the idea of a Precambrian ultrametamorphic series in the Central Rhodope Mts.

Introduction

The results of the radiogeochronological research in the Rhodope crystalline complex during the last 10-15 years have questioned a number of points in the tectonic concept treating the structure and evolution of the so-called Rhodope medium mass in terms of Precambrian processes of formation and consolidation. The ideas that two metamorphic complexes of different ages, Archean and Proterozoic, are present in the Rhodope Mts and that the "Southbulgarian granites" in the Rhodope area are either of Precambrian or of Paleozoic age have not been corroborated by radioisotope geochronology so far (Йорданов et al., 1962; Арнаудов et al., 1969, 1974, 1979, 1980, 1988, 1990; Amov et al., 1973, 1982; Пальшин et al., 1974; Бояджиев & Лилов, 1976; Загорчев & Мурбат, 1986). Most Bulgarian geologists studying the high-metamorphic series and the granitoids emplaced in them regard the Phanerozoic age values and especially the younger, Alpine ones gained for rocks from the Southbulgarian crystalline sequences both in the Rhodope and in the Srednogorie zones either as totally erroneous, or as derived by radiogeochronological methods ill-suited for the purpose, or as reflecting late processes of alteration superimposed on the crystalline massif. These values are rejected as representing the real ages of the original metamorphic and magmatic events, of the introduction of fresh matter into the area.

This communication is concerned with just such young age values derived by the U-Pb method for zircons from migmatic pegmatites in the Central Rhodope metamorphic complex generally believed of Precambrian age.

Review of published work

Костов (1940) was the first to report on migmatic pegmatites in the Rhodope crystalline complex in his study of allanite from pegmatite interlayers in the metamorphic rocks near the village of Mihalkovo. Later on, Вергилов (1959) has studied the structure, composition and origin of migmatic pegmatites crosscutting marbles in the same area. Арнаудов & Петрусенко (1967) have described migmatic pegmatites with moissanite and allanite. The distribution and the relationships of bedded migmatic pegmatoid veins with later cross pegmatites have been the subject of a number of papers (Вергилов, 1960; Вергилов et al., 1963; Кожухарова & Кожухаров, 1962; Боянов et al., 1963; Кожухаров, 1968, 1971). Later studies of migmatic pegmatites concentrate on the geochemistry of some trace elements as related to the metamorphic and pegmatite-forming processes (И. Иванов et al., 1984, 1989; Алексиев et al., 1987), and on the reconstruction of syn- and postmetamorphic mineralization events in the Central Rhodope area (Костов et al., 1986).

Geological background

The migmatic pegmatites studied occur in a series of biotite, amphibole-biotite gneisses and marbles exposed along the Krichim—Devin road near the Antonivanovtsi dam lake. In the lithostratigraphic scheme of Кожухаров (1984), these metamorphic rocks belong to the Vacha Formation of the Rupčos Group in the Rhodope Supergroup and are of Proterozoic age. In the scheme of Ж. Иванов et al. (1984), the same rocks are referred to the Posestrimo Formation of the Chernatitsa Group.

The migmatic pegmatites are mostly bedded to low-angle cross sheets, the thinner veins commonly having diffuse boundaries with the host migmatic gneisses. Their contacts with the host amphibolites and marbles are sharp. Thicknesses vary from several centimeters to 1 or 2 m, lengths reaching several meters. Occasionally, there are veins up to several meters thick and 40-50 m long (И. Иванов, 1989). They show incipient zonal structures of granitic, aplitoid and pegmatoid zones (И. Иванов, 1989). Blocky quartz-feldspar zones have also been observed in individual pegmatite veins, in their central parts mostly.

The main rock-forming mineral constituents of a vein and their proportions are largely determined by the composition of host metamorphic rocks. Bedded vein pegmatite formations up to several centimeters thin, not always reliably discernible from the eyed-banded leucosome of the host migmatic gneisses, show a comparatively simple mineral composition of feldspar (plagioclase$\pm$potassic feldspar), quartz$\pm$biotite, magnetite, apatite, zircon$\pm$ allanite. Thicker and better defined bedded to low-angle cross pegmatite veins additionally contain amphibole, garnet, sphene, rutile, epidote and pyrite. Individual veins have been found to contain kyanite and moissanite as well (specimens of Stancheva and Cherneva). In the thick cross pegmatites, garnet and sphene become more abundant, and muscovite and occasionally tourmaline appear. The mineral paragenesis most typical of the bedded migmatic pegmatites in the area between the town of Krichim and the Antonivanovtsi dam lake consists of allanite (up to 1.5%) and zircon (up to 0.1%) (И. Иванов, 1989).

Description of zircon

For the purpose of this study, we had extracted zircon crystals from migmatic pegmatites and gneisses south of Krichim and examined their morphology and distribution of morphological types. Although still insufficient, these observations have permitted us to draw some preliminary conclusions relevant to our interpretation of radiogeochronological results.

Zircon is present in all pegmatite formations but its content is higher in the bedded to low-angle cross pegmatites enriched in allanite, consisting of orthoclase and plagioclase, occasionally of plagioclase only, and occurring in biotite and amphibole-biotite gneisses, or in marbles; zircon is less abundant in the well defined cross veins with sharp contacts.

In the bedded migmatic pegmatites from the area of the Antonivanovtsi dam lake, zircon occurs as well formed, sometimes slightly rounded, pale-pink to pink crystals. Metamict zircons have not been observed, and cloudy or cracked crystals are very rare. Dimensions vary widely, from 0.1×0.05 mm to 4-5×1-2 mm. Colour is deeper in the larger crystals which fairly often contain inclusions of biotite, allanite and sphene (?).

The following morphological types of zircon have been observed in the pegmatite veins Y-1501 and Y-1545 from which crystals for radioisotope analysis were extracted:*

— nearly isometric crystals resembling garnet, small (0.25-0.6×0.2-0.5 mm), pale-pink to pink, commonly rounded, of slightly drop-like shape. They are bounded by unevenly developed {110}>{100}, {211} and {101}. The most prominent forms usually are {211} and {110}. Frequency of occurrence 5-16%;

— relatively large (0.7-1.2×0.35-0.5 mm) well formed pink crystals of elongation factor 2. Habit form {211}, subordinate forms {100}≤{110}, small {111}, {101} and {112}. Frequency of occurrence 35-44%;

— prismatic pink crystals 0.5-1×0.3-0.45 mm large of elongation factors 2.2-2.7. They are bounded by {100}>{110}, {211}, {101} and {112}. Frequency of occurrence 20-25%;

— long-prismatic pink crystals 0.7-1×0.2-0.3 mm large of elongation factor 3, bounded by {100}={110}, {211}>{112}≥{101}, habit forms {100} and {110}. Frequency of occurrence 12-15%;

— markedly prismatic pink crystals 0.6-1.2×0.25-0.4 mm large of elongation factors 2.5-3. Prisms predominate with {100}≫{110}; {211} and especially {101} are very small. Terminals are usually rounded. Frequency of occurrence 5-10%.

In the later, thicker, well defined cross pegmatite veins and bodies whose mineral assemblage expands to include muscovite, garnet, rutile, kyanite and tourmaline as well, zircon crystals are mostly small (0.15-0.5×0.02-0.3 mm), prismatic to long-prismatic, colourless or wax-yellow to amber, occasionally perfectly clear, sometimes poorly formed with curved edges and strongly rounded terminals. There are also distorted subparallel aggregates growing from narrow bases and splitting towards the terminal pyramidal zones. Faces show grainy texture. Habit forms {110} and {101}; small {100} and {211}, strongly rounded. There are very rare spear-shaped crystals with well-formed {211}. The length-to-width ratio varies between 2 and 10; elongation factors 4-8 are most common. Flat, usually cracked, clear crystals of highly asymmetric development of {101} have also been observed in some pegmatite veins.

Zircon crystals from the host gneisses of bedded and cross pegmatites are also very small (0.1-0.6×0.05-0.3 mm), colourless to very pale pink, occasionally cloudy and of rounded pyramidal terminals. They show markedly prismatic habits, the elongation factors being 2.5-3, rarely up to 5. {110} commonly is the predominant prism with {100} usually subordinate. {101} is always present, and {211} and {301} are very infrequent and markedly smaller than {101}. Short-prismatic zircon crystals of elongation factors 1.5-2 occur in

* 20 goniometric measurements of φ and ρ made by I. Vesselinov on faces belonging to 3 simple forms on 4 crystals gave a mean c/a=0.9037 with σ=0.0040; compare to c_0/a_0=0,9014 in Kostov (1968). The crystallographic orientation of zircon in this study follows Kostov (1968).

some eyed-banded biotite and amphibole-biotite gneisses. They show more pronounced development of dipyramidal faces, chiefly {211} with {101} being subordinate. Pink colour becomes deeper.

Radioisotope data

Isotope analyses for the purposes of radiogeochronology were made in two laboratories, one in Sofia and the other in Amsterdam, Holland.

In 1978, in the Laboratory of Mass Spectrometry at the Institute of Nuclear Research Bulgarian Academy of Sciences, B. Amov determined the age of zircon Y-1501 sampled from a bedded pegmatite vein in migmatic biotite gneisses near the Antonivanovtsi dam south of the town of Krichim. Uranium and lead were analyzed in aliquot samples after $NaHF_2$ decomposition of zircon (П а в л о в а, 1978). Uranium, extracted with TOPO in 2n HNO_3, was analyzed by spectrophotometry using PAN, and lead was determined by isotopic dilution following a dithyzone extraction. Isotope analyses were made with a modified MU-1305 mass spectrometer. Its low sensitivity required relatively high lead concentrations (>10 ppm) for isotope analysis. The zircon sample separated for the radiogeochronological study was too small (about 0.4 g) to permit analysis of lead isotopes in various zircon fractions defined by grain-size or some other physical characteristics because of which the whole lead yield gained from the available zircon sample (about 34 ppm) was analyzed. The correction for ordinary lead was derived from isotope analyses of lead in potassic feldspars from Alpine granitoids and pegmatites in the Rhodope Mts.

In 1980, Ts. Baldjieva under the guidance of Dr. E. Hebeda determined the age of zircon Y-1545 in the ZWO Laboratorium voor Isotopen in Amsterdam. The zircon Y-1545 sample had been derived from bedded pegmatites occurring in migmatic gneisses near the Krichim Spa. Two fractions were analyzed: 1 — rounded zircon crystals, and 2 — crushed angular ones. Chemical decomposition and lead extraction followed chiefly the method of K r o g h (1973). Anode deposition was used for lead separation (A r d e n & G a l e, 1974). Isotope rations were measured on a computer-controlled Teledyne SS 1290 mass spectrometer with a Faraday cylinder as collector and a digital interface.

The age values derived in the two laboratories from the $^{206}Pb/^{238}U$ and $^{207}Pb/^{235}U$ ratios are very close to each other (Table 1). The results obtained in the Amsterdam laboratory for the two zircon Y-1545 fractions show practically no differences either, which is another argument in favour both of the real validity of the ages determined and of the low probability that zircons of appreciably different ages of formation were present in the analyzed samples. If some, even very low amounts of old, Precambrian zircons were present in the migmatic pegmatites, they would have changed perceptibly the U-Pb isotope ratios, and accordingly the calculated ages. The physical characteristics of zircon crystals, in spite of the several habit types observed, are also incompatible with an assumption that would consider different zircon generations far removed in time of formation.

T a b l e 1

Zircon ages

No of sample	U ppm	Pb ppm	Observed atomic ratios			Atomic ratios		Apparent ages (Ma)	
			$^{206}Pb/^{204}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{235}U$	$^{206}Pb/^{238}U$	$^{206}Pb/^{238}U$	$^{207}Pb/^{235}U$
Y-1545									
rounded crystals	157	1.52	775	0.06918	0.2303	0.05893	0.008453	54.3	58.1
crushed crystals	163	1.59	634	0.07167	0.2420	0.05629	0.008392	53.9	55.6
Y-1501	157	34	21.06	0.7502	1.8623	0.05189	0.007661	49±5	51±10

Discussion

According to the views of some authors (Боянов et al., 1963; Кожухаров, 1968, 1971), the Rhodope metamorphic sequence hosts several types of pegmatites differing in age and genesis and including migmatic, mostly bedded pegmatites associated with ultrametamorphic processes during both the Archean and the Proterozoic, and cross pegmatites of poorly understood origin. It has been suggested that the cross pegmatites are residual products of a Paleozoic igneous activity (Бояджиев, 1959; Боянов et al., 1963; Вергилов et al., 1963; Кожухаров, 1971), and some of them in the Mihalkovo area, Central Rhodope Mts, even of a Laramide magmatism (Кожухаров, 1968). Migmatization and granitization have been regarded as the result of injection of bedded migmatic pegmatites which converted the gneisses into banded and ribbon migmatites (Боянов et al., 1963; Вергилов et al., 1963; Кожухаров, 1968, 1971). It is believed that the Prerhodope sequence suffered migmatization three times whereas the Rhodope one only once (Кожухаров, 1968, 1971; Кожухарова & Кожухаров, 1980). The last of the three stages of ultrametamorphism (the Archean-Proterozoic boundary) coincides in time with the regional metamorphism of the Rhodope complex (Кожухарова & Кожухаров, 1980). According to this scheme, the bedded migmatic pegmatites in the Vacha Formation should have been formed during exactly that stage.

The age values derived from the two zircon samples define a Late Phanerozoic, Tertiary time of formation for the respective type of pegmatite formations. It follows that the cross pegmatite veins in the so-called Vacha Formation are even less referable to the Precambrian or Paleozoic.

The young ages of the Central Rhodope bedded migmatic pegmatites are not surprising. Similar age values, 53-49$\pm$5 Ma ($^{206}Pb/^{238}U$) and 53-49$\pm$8 Ma ($^{207}Pb/^{235}U$), were obtained about 20 years ago for uraninites from pegmatite veins crosscutting biotite gneisses near the village of Dolen, Zlatograd district, in the Eastern Rhodope Mts (Арнаудов et al., 1969). Zircons (zircon+cyrtolite) from the same pegmatites showed ages of 38$\pm$15 Ma ($^{206}Pb/^{238}U$) and 40$\pm$15 Ma ($^{207}Pb/^{235}U$).

Analogies are not proofs of similar genesis, or age, yet we are tempted to point out the similarity between the pegmatites near the village of Dolen and the cross pegmatite veins occurring in the gneisses of the variegated formations in the so-called Rupchos Group near Chepelare and the village of Bogutevo. In both localities, near Dolen and near Chepelare, the cross pegmatites are largely of the oligoclase-microcline and microcline-oligoclase paragenetic types (potassic feldspar is mostly orthoclase) with muscovite, biotite, garnet, kyanite, zircon$\pm$cyrtolite, apatite, monazite, xenotime, allanite, niobotantalates (columbite, yttrotantalite, struverite, tanteuxenite) $\pm$ beryl (Иванов & Арнаудов, 1966; Арнаудов & Петрусенко, 1971, 1972).

Among the cross oligoclase-microcline pegmatites in the Rupchos group there are also pegmatites of less rich accessory mineralization represented by garnet, sphene, allanite, apatite, zircon, kyanite $\pm$ zircon which makes them compositionally similar to the bedded migmatic pegmatites.

To the above comparison of mineral compositions we may add the trends of morphological modification and chemical variation in zircons from the respective pegmatites. The appearance of cyrtolite in the cross pegmatites, accompanied by increased U and Th contents, and especially the steady rise of hafnium contents from the zircons in the bedded pegmatites towards the zircons in the zonal, cross pegmatites (Апостолов et al., 1975) are also compatible with the idea of a genetic relationship between the bedded and a major part of the cross pegmatites. Further arguments in favour of a common origin of the migmatic gneisses, the bedded and cross pegmatites, all products of migmatization processes, are supplied by the data on the geochemical behaviour of a number of trace elements (И. Иванов et al., 1984; Чернева et al., 1986, 1987, and unpublished data of the same authors). The transition from bedded to cross pegmatites is characterized by a directed increase of

Rb, Be, Nb and Ta contents, and a decrease of TR, Ba, Sr, Zr contents and of Ba/Rb and Rb/Sr ratios (И. Иванов et al., 1984).

The evolution of mineral and chemical compositions from the more primitive bedded formations to the cross (of sharp contacts with the host gneisses) migmatic pegmatites from the Vâcha Formation finds its plausible explanation in the results of the thermobaric studies and paragenetic analyses of the Central Rohodope syn- and postmetamorphic mineralizations (Костов et al., 1986). That interpretation assumes that the first stage of the so-called anatectic-pegmatite phase, which followed a phase of prograde metamorphism, produced injection gneisses, banded gneisses and granite gneisses formed in the stability fields of sillimanite and partly of kyanite. The second stage of that phase produced the bedded to low-angle cross injection migmatic pegmatites as the result of mild tectonic acton, and the final stage, involving uplifting movements and consolidation of the metamorphic sequence as wel- as temperature lowering, yielded the typical cross migmatic pegmatite veins formed by residual anatectic fluids (Костов et al., 1986).

Another study corroborating a single-phase process of migmatization and pegmatite formation again within the time span of the Alpine tectogenesis is the isotope analysis of lead in metamorphic rocks from the so-called Lyubinovo Formation in the Arda Group of the Prerhodope Super-group (stratigraphy after Кожухаров, 1984). The model ages (65-35 Ma) derived from the isotope ratios of lead in potassic feldspars from various migmatic formations exposed along the Egri Dere valley north of the town of Ardino (Арнаудов et al., 1990) are incompatible with the ideas defining several migmatization phases in the Central Rhodope area which took place during the Archean and at the Archean-Proterozoic boundary. The U-Pb age calculated by Bartnitsky for zircons from migmatic gneisses in the Lyubinovo Formation shows analogous values (58-33 Ma) (Арнаудов et al., 1986, 1990). It is obvious that the ages derived by the U-Pb method as well as the model ages based on the lead isotopes date one and the same process of ultrametamorphism in the Central Rhodope crystalline complex.

The manifestations of an Alpine migmatization question not only the almost generally accepted and popular concepts on the chronology of metamorphic events in the Central Rhodope area but also the lithostratigraphic schemes and notions on the tectonic evolution of the entire crystalline complex in the Rhodope zone. Radiogeochronology has found so far no evidence sustaining the notion of a Precambrian regional metamorphism and ultrametamorphism. The oldest radioisotope age values derived for Central Rhodope metamorphic rocks remain within the time span of the Phanerozoic. They have been obtained for amphibole-biotite gneisses near the village of Shiroka Lâka belonging to the so-called Sakar Group (A_2) of the Prerhodope Supergroup according to the lithostratigraphic scheme of Кожухаров (1984). The U-Pb age of zircons from these migmatized metamorphic rocks, as derived by Bartnitsky (in: Арнаудов et al., 1986)) falls within the range of 242-210 Ma. The geochemical study of Чернева et al. (1987) has proven the orthogenesis of those metamorphic rocks. The interpretation advanced by these authors considers the formation of several facies varieties of granitoids, ranging from amphibole-biotite to aplitoid granites, as the result of an intrusion and subsequent differentiation of granite magma. A regional metamorphism into amphibolite facies converted the granitoids into gneisses which were later migmatized. The mineralogical characteristics of zircon in these gneisses have provided evidence of their magmatic origin. Most likely, the calculated U-Pb age values fix the time of granite formation, whereas the superimposed metamorphic as well as ultrametamorphic changes during the Alpine times have left no detectable traces in the uranium-lead isotope system in zircons of the orthogneisses.*

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* Note of Editorial Board. The interpretations in this paper are subject of discussion because not taking into consideration the presence in the same area of non-metamorphosed Paleogence formations (biostratigraphically proven Paleocene, Upper Eocene and Lower Oligocene) which cover unconformably the Rhodope crystalline complex.

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